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## (54) TOBACCO CONTAINING ENCAPSULATED FLAVOR

(71) We, INTERNATIONAL FLAVORS AND FRAGRANCES INC., a Corporation organised under the laws of the State of New York, United States of America, of 521 West 57th Street, New York, New York, United States of America, do hereby declare the invention for which we pray that a patent may be granted to us and the method by which it is to be performed to be particularly described in and by the following statement:—

This invention relates to an improvement in tobacco whereby a plurality of minute, heat rupturable capsules each containing a synthetic clove oil as an inner phase are mixed in integral contiguous relationship with shredded tobacco. As tobacco surrounding each discrete capsule burns, the elevation in temperature is such that capsule walls rupture, thereby releasing a discrete quantity of clove oil vapor which becomes entrained in smoke issuing from the burning tobacco. The capsules being spaced homogeneously throughout the length of the smoking article, the capsule walls rupture successively, or consecutively, with the advance of the burning front of ignited tobacco. The exscent smoke inhaled by the smoker is homogeneously aromatic. The capsules are of such a size (from 50—500 microns in diameter) that the rupture thereof gives rise to an audible crackling sound; a property desirably for cigarettes, cigars, etc. marketed in certain nations in the Far East such as Indonesia.

This invention also relates to shredded tobacco and more particularly to capsule-containing shredded tobacco compositions, methods for producing the same, and to smoking articles, such as cigarettes, cigars and the like containing desired quantities of encapsulated synthetic clove oil.

The present invention can be practiced by admixing a capsule slurry with shredded tobacco. It has been found that although tobacco tends to burn with acidity, the

acridity is masked or nullified by the flavoring of smoke from the burning of said tobacco with encapsulated odoriferous clove flavors.

It has been recognized that the addition of clove oil as well as other tobacco flavor enhancers to tobacco imparts a pleasant flavor to the tobacco. In methods hitherto used, the tobacco has been treated by soaking it in a suitable solution of natural or synthetic clove oil or the solution has been sprayed on the tobacco. One disadvantage of such processes is that the clove oils which are suitable for this use have an appreciable vapor pressure at ordinary temperatures and for that reason the substance more or less rapidly vaporizes and escapes. Furthermore, natural clove oils are relatively expensive. In the process of this invention, inexpensive synthetic clove oil may be used and when encapsulated in microcapsules, this substance does not vaporize and escape. In addition, the microcapsules are of such a size (50—500 microns) that when the smoking article is burned, when in use, the capsules rupture with an audible crackling sound. Such a property is desirable in smoking articles marketed in such Far Eastern nations as Indonesia. A means of overcoming the acridity of shredded tobacco by including synthetic clove oil containing capsules admixed therewith is to mix the capsules in slurry form with the shredded tobacco.

In addition, the subject invention provides a system for the penetration of capsular clove oil-containing material applied to a wet fibrous tobacco sheet which affords the advantages inherent in having said capsules disposed within the body of the sheet material and yet accomplishes this end without concomitant intolerable loss of the capsules to achieve it. The sheet is shredded subsequent to being coated and dried.

In the tobacco industry, and particularly that part of the industry relating to the manufacture of cigarettes, cigars and like

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smoking articles, large quantities of tobacco are rejected annually and sold as waste because it is either impossible or impracticable to utilize them in the manufacture of smoking articles. Obviously the loss of these materials is a serious matter from the point of view of the manufacturers and, therefore, it would be highly desirable if such materials could be salvaged and used in the manufacture of smoking articles. Materials now disposed of as waste are tobacco stems, dust, scrap, clippings and other portions which are unsatisfactory in connection with the manufacture of high-grade smoking articles.

From time to time attempts have been made to form tobacco sheets or films which would solve the problem of satisfactorily disposing of or utilizing tobacco waste materials. Processes have been developed for forming tobacco paper utilizing well-known paper processes and techniques in which tobacco waste, such as for example stems and scraps, has been reduced to pulp by the use of chemicals, and then according to ordinary paper making techniques converted into paper. These techniques are unsuitable for use in connection with the present invention in forming improved tobacco sheet material, carrying out the processes therefor, and producing smoking articles therewith, such as cigarettes or cigars, because of the radical changes to which these tobacco materials are subjected, and the loss of valuable constituents and properties of natural tobacco.

According to one aspect of this invention, tobacco sheet or film material may be made along lines similar to those described and disclosed in U.S. Patent 2,433,877, in which the final product contains substantially all of the desirable properties and natural characteristics of tobacco, especially insofar as aroma, taste and color are concerned, and has the same general appearance of tobacco.

An object of this invention is to provide an aromatic content which is closely similar to distilled clove oil to smoke from an unfiltered cigarette, or a filtered cigarette.

Another object of this invention is to prevent the premature loss of volatile synthetic clove oil flavoring material from smoking articles such as cigars and cigarettes.

Another object of this invention is to coat reconstituted tobacco sheet material with capsules containing a synthetic clove oil material.

Another object of this invention is to provide a synthetic clove oil flavor containing reconstituted tobacco sheet material in which the clove oil flavor is invisible and undetectable prior to smoking said reconstituted tobacco sheet material.

Another object of this invention is to provide a mixture of shredded tobacco and microcapsules which yield a volatilized clove oil flavor when the tobacco is burned, and,

simultaneously rupture with an audible crackling sound.

Another object of this invention is to provide a reconstituted tobacco sheet which has the advantage of physical protection of capsules containing synthetic clove oil flavoring from touch and sight. To the end that the particles (capsules may be confined within the sheet, they are of necessity smaller in their average diameter size than the sheet thickness and preferably are of such size, and present in such quantity, that they are protectively nested in the interstitial spaces between the tobacco fibers of the reconstituted tobacco support sheet, in addition to being adherent to the tobacco fiber, even if calendered, without disturbing, from a user's viewpoint, the normal physical structure of such a sheet made without them.

The present invention encompasses mixtures of shredded tobacco and microscopic capsules each containing an inner phase comprising volatile flavoring material such as a synthetic clove oil material; and methods of forming such mixtures, including the method of forming continuous, self-supporting reconstituted tobacco films or sheets which comprises forming an aqueous slurry of comminuted tobacco stems, forming said slurry into a continuous thin web, and applying to said web microscopic capsules each containing an inner phase comprising volatile clove oil flavors and, if desired, additional tobacco flavor enhancers such as those described in Canadian Patents 805,933; 805,934; 805,459; and 805,460.

The tobacco stems, veins, and tobacco leaf material can be comminuted by means of a suitable type of mill, such as a ball mill or colloid mill. Any desired ratio of tobacco material to water can be used. It has been found that ratios of 1:8 to 1:14 give satisfactory results. When the milling or slurry forming operation is completed, the slurry is deposited in a film forming device and applied in the form of a continuous film or web upon a suitable film forming surface, such that when the slurry is spread out in the form of a film or web thereon, substantially all soluble and insoluble constituents of the tobacco stems and veins will be contained in a reconstituted tobacco sheet or film. The tobacco film can be coated with encapsulated clove oil flavoring material while in either wet or dry condition. If desired, on drying, the resulting web may be shredded.

Alternatively, this invention encompasses the mixing of previously shredded tobacco with capsule slurries whereby the individual capsules adhere to shreds of tobacco.

The present invention provides means for making the aromatic synthetic clove oil substance constantly available in a fresh condition by encasing it in an easily rupturable

capsule located in contiguous contact with tobacco.

It is neither necessary nor desirable to rupture the capsules contained in a tobacco smoking article since heat from a burning front of tobacco will rupture the capsules serially thereby releasing aromatic material such that (1) the smoke emanating from the tobacco smoking article is homogeneously flavored with the synthetic clove oil and, if desired, other tobacco flavor enhancers such as 4 - hydroxy - 4 - methyl - 5 - hexenoic acid,  $\gamma$  - lactone described in Canadian Patent 805,934, and (2) a sound audible to the human ear is given off at the instant of rupture.

One of the features of the instant invention is the saving in volatile synthetic clove oil material effected by encapsulation. The evanescence of the volatile materials is greatly inhibited by the intact capsule wall.

This invention includes the application of one or more other sets of capsules throughout shredded tobacco capsule mixtures; and also throughout the thickness of the reconstituted tobacco sheet or on the outside of the sheet, and it does not preclude the presence of other material, particulate or other, present in, on the outside of, or otherwise associated with the shredded tobacco mixture or tobacco sheet, to co-operate, or not, with the set of capsules positioned in the sheet by the practice of this invention cumulatively or countervailing, which produce a beneficial effect. There may be two or more sets of capsules in the mixture or sheet being confined or not confined to the surface of the tobacco shreds or sheets or spread throughout the thickness of the sheet or discontinuously through the tobacco mixture.

It is contemplated that the capsules need not be of uniform substance, content, and size so long as their diameter is in the range of 50—500 microns. The capsules may be of mixed sizes and wall materials, or of different content or origin, in order to serve one or more utilitarian purposes in behavior, or to have walls of varying resistance to rupture or fracture, or to be different in any other aspect of structure or performance (such as resistance to heat) or different in any olfactory sense characteristics—or to contain, in addition to the synthetic clove oil, chemical reactants, medicines, perfumes—absorbents or adsorbents—as between the fractured and unfractured state or in either state—or to be affected by other forms of energy in various manners which will be evident to those skilled in the art. Each of the capsules may be self-contained, in that each, upon rupture, releases its volatile content including synthetic clove oil. The particulate liquid may contain either liquid or solid solute material dissolved therein, or have such materials dispersed therein in any physical state, and may con-

tain materials which may be subject to growth, metamorphosis, or degeneration.

To obtain disuniformity among capsules if such are used, they may be manufactured in separate batches according to the characteristics of the capsules as to origin, size, contents, or wall material and structure, and thereafter blended in the desired proportions for introduction into the paper as a single applied slurry. This is important where more than one kind of capsule contents (solid or fluid) is to be made use of either by rupture of the capsules or by their characteristics in the unruptured state. It is within the province of this invention to provide capsules having different thicknesses of walls or different strengths of walls, to the end that they may be differentially fractured by variations in temperature, so that some capsules may be ruptured by one temperature and the rupture of other capsules will follow in response to application of a greater temperature.

In a preferred embodiment, though not limited thereto, a system is capable of limiting the penetration of capsules containing synthetic clove oil into a shredded tobacco mixture or into a reconstituted tobacco sheet as the sheet is being formed, so that the particles are substantially concentrated subsident to either surface of each of the shreds or of the sheet in a firm, well defined stratum of binder material.

The capsules may be applied to shredded tobacco or tobacco sheet in an aqueous slurry containing a solvent-responsive or solvent-activated adhesive such as a vegetable gum or starch. It is to be understood that the use of such adhesive is not necessary to the operation of this invention. Efficacious natural adhesives or binders include hydrolyzed starches (principally tapioca starch), and natural gums such as gum arabic, gum tragacanth and karaya gum. These adhesives or binders are used exclusively in water solution. (While karaya gum is not actually soluble in water, karaya gum particles absorb large amounts of water and swell to great size). Certain water soluble synthetic adhesives or binders such as polyvinyl alcohol, sodium cellulose glycolate, and methyl cellulose can be used to bind capsules onto tobacco shreds or in a reconstituted tobacco web.

The capsules preferably should contain an oily, aromatic liquid comprising about 85%, by weight, of the encapsulated structure and containing synthetic clove oil; and, if desired, other tobacco flavor enhancers. The oily liquid is released by the rupturing of the capsule wall material. The oily liquid may be a mixture of oils or dispersions of solids in oils containing, inter alia,  $\alpha$ ,  $\beta$  and  $\gamma$  caryophyllene; furfural; eugenol; acetyl eugenol; and eugenyl acetate. The oils can also include furan, methyl furan, 1 - hexene, cyclo-

pentadiene, benzene, cyclohexane, and other sesquiterpenes having molecular weights in the range of 204 up to 240. The aggregation of capsules can contain disparate substances within each discrete capsule.

Preferably, from about 60 up to 80% of the oil is eugenol; with the remainder of the oil comprising the other aforementioned ingredients.

This invention also includes smoking articles containing two disparate encapsulated substances which act synergistically when the capsules containing them are ruptured. The relative proportions of solid particles to capsules may vary according to the effect desired.

The capsule walls can be made of a gelatin-gum arabic complex, gelatin-carrageenan complexes, phased out succinylated gelatins, ethyl cellulose, or other material. Any standard method of encapsulation such as the one set forth in Example I may be used to form the capsules.

The size of the capsules can range from approximately 50 microns in diameter to approximately 500 microns in diameter, and may be mixed as to size, or even mixed as to content. Capsules spaced in the cigarette should not exceed 500 microns in diameter and should not be less than 50 microns, since smaller capsules fail to rupture with a report loud enough to be desirable, and larger capsules may separate too easily from the wet tobacco and from the smoking article itself.

The weight ratio of dry tobacco to dry capsules may be in the range of 1:0.01 up to 1:0.08 with the preferred range being from 1:0.02 up to 1:0.04.

Capsules of the 50 to 500 micron size can be made according to the process disclosed in United States Patent No. 2,800,457, which issued on July 23, 1957, to Barrett K. Green and Lowell Schleicher. Briefly, that patent discloses film-forming polymeric materials of opposite electric charge in aqueous solution which are caused to form a separate liquid dispersed phase (a coacervate) by modifying the conditions of the solution.

The intended oil contents of the capsules containing synthetic clove oil are emulsified in the solution, and the coacervate forms a liquid film about the emulsion. The film is made firm and solid, and is then hardened. The capsules are recovered ready for their use as part of the heat rupturable composition. The oily material of the preferred embodiment (e.g., a solution containing eugenol) may be modified by addition thereto of the previously mentioned or other materials of aromatic or therapeutic properties.

The reconstituted sheet which is made for use in this invention, can be used either in the manufacture of cigars, cigarettes or other like smoking articles.

In the following description, no distinction is made between the types of tobacco employed.

In making sheet or film material to be expanded and made available for use in the manufacture of smoking articles, a suitable slurry can be formed from seventy-five per cent stems, which are run through a conventional comminuting machine in order to reduce the stems to extremely fine size after which they are mixed with water, and twenty-five per cent tobacco scrap leaf portions, dust or fines which have also been passed through the comminuting machine and preferably screened to remove any foreign material such as sand mixed therewith. The mixture of comminuted stems and leaves is then placed in a ball mill of known design, and ball milled for a period of time, preferably sufficient to completely hydrate or gelatinize the particles of tobacco being ball milled. The size of the ball mill and balls, and speed of rotation, control and amount of time necessary to reach the desired state of hydration or gelatinization of the tobacco particles. For instance, it has been found that in a four gallon jar ball mill using three-fourths inch nominal size flint pebbles, the ball milling time may approximate from six to sixteen hours at fifty R.P.M. whereas in a five quart ball mill using the same size pebbles at sixty R.P.M., the minimum time may run as high as thirteen hours.

Upon completion of the slurry, it is removed from the ball mill and placed in a film forming device and formed into a continuous sheet or web. The sheet or web material is then ready for the addition of capsules in a slurry.

A primary objective of the invention is to provide a reconstituted tobacco sheet or shreds with limited penetration of liquid-containing capsules as the particles that, because of their position in the sheet or on the shreds, when ruptured by heat, release the contained volatile clove oil flavoring liquid. This objective is accomplished by forming a sheet of reconstituted tobacco with the capsules embedded in it or by forming a mixture of tobacco shreds to which have been applied capsules which are adhered thereto or embedded therein. The amount of capsules applied should be related to the final sheet or shred thickness and the amount of encapsulated liquid necessary or desirable to be provided, and the average shred width.

In the application of the subject invention to the production of sheet material, the preferred system comprises laying down a wet web of tobacco fibrous material on a supporting member to form an embryonic sheet, and applying to said web minute capsules, the contents of which capsules comprise flavoring material.

Following the formation of the sheet of

web, it is moved through a moisture removing zone and excess moisture is removed therefrom.

It is within the scope of this invention to add the capsules by any means at any point in the tobacco web formation, provided that the web at the time of addition of the capsules is sufficiently wet and penetrable, either by reason of undrained liquid or because the capsular particulate material was added as a liquid slurry, to permit fixation on the tobacco fibers. Furthermore, it is within the scope of this invention to add the capsules to the shredded tobacco at any point in the shredding operation so that the capsules are not ruptured.

Following are examples of systems that produce either a controlled penetration of capsules within a reconstituted tobacco sheet or web; or produce a mixture of shredded tobacco and capsules adhered to the tobacco shreds:

#### EXAMPLE I

The following is a description of a preferred embodiment of the invention as carried out using a process wherein minute capsules having a diameter in the range of 50 up to 500 microns were added to a wet web of reconstituted tobacco (weight ratio of dry web to dry capsules=1:0.04. The capsules and binder materials (weight ratio of dry capsule to dry binder=1:0.1) when placed among the tobacco fibers, wet them and entangle with them and clothe them, thus in effect securing and binding the capsules against migration through the sheet, thereby forming a subsident stratum. The majority of binder and associated capsules are caught in the sheet. Substantially no capsules migrate through the sheet. When the wet tobacco web is dried, the binder shrinks by loss of solvent, leaving the dried polymeric binder material, and the capsules remain in place relatively with respect to sheet thickness. The sheets containing the capsules are then shredded and used in producing smoking articles such as cigarettes. Such cigarettes are formed using a wrapper, containing a fill of tobacco extending from one end of the wrapper to the other, and intimately admixed with the tobacco, a plurality of microcapsules each comprising an aromatic volatile synthetic clove oil flavorant. The capsules are homogeneously spaced in contiguous relationship with the tobacco such that as the burning front of the tobacco advances the length of the tobacco article, a concomitant elevation of temperature initiates consecutive rupture of the capsules (1) releasing the volatile synthetic clove oil-containing material which emanates with smoke from the smoking article into the smoker's mouth and (2) yielding a crackling sound audible to the human ear.

Tragacanth gum solution and starch solution were prepared for use as alternative binders in the following manner:

#### Part A

##### Concentrated Tragacanth Gum Solution (binder)

4.5 pounds of dry tragacanth gum powder was stirred into 50 gallons of water, using a suitable mixer. Five minutes after all the powder had been added, the mixer was turned off. The tragacanth gum solution was allowed to sit for two hours, and then the mixer was turned on for five minutes. Sitting for two additional hours, enabled the tragacanth gum to hydrate. After five minutes, the mixer was turned off, and the 55-gallon drum was covered. Just prior to combining the tragacanth gum solution and the capsular slurry, 50 gallons of tragacanth gum solution was diluted with water to 3% tragacanth gum on a solids basis.

#### Part B

##### Starch Solution

The hydrolyzed starch solution was prepared by heating a slurry of the starch at 195 degrees Fahrenheit for a minimum of fifteen minutes to provide a 1%, by weight, starch-in-water solution.

Capsule slurries were prepared in the following manner:

#### Part C

##### Preparation of Synthetic Clove Oil

A mixture was prepared containing the following ingredients:

| Ingredient              | Percentage |     |
|-------------------------|------------|-----|
| $\alpha$ -Caryophyllene | 2.0        | 100 |
| $\beta$ -Caryophyllene  | 2.0        |     |
| $\gamma$ -Caryophyllene | 10.0       |     |
| Furfural                | 0.1        |     |
| Eugenol                 | 75.0       |     |
| Eugenyl Acetate         | 5.0        | 105 |
| Acetyl Eugenols         | 5.9        |     |

#### Part D

##### Preparation of Capsular Slurry-Encapsulation of Synthetic Clove Oil

10 grams of gum arabic were dissolved at room temperature in 220 grams of deionized water. The mixture was agitated until the gum arabic was fully dissolved. In a separate 250 milliliter Erlenmeyer flask, 10.0 grams of modified gelatin was mixed with 220.0 grams of deionized water. The gelatin was allowed to tumefy at room temperature; and also then warmed in a water bath to about 40°C. with stirring so that the gelatin was dissolved.

The gelatin solution and the gum arabic solution were poured into a beaker equipped with a stirrer. A flocculence indicating the precipitation of the gelatin was noted. The

temperature of the mixture was decreased to 35°C. The speed of the stirrer was adjusted so that it was turning only enough to keep the phases mixed. The pH of the mixture was 4.50.

Into the beaker containing the mixture of gum arabic and gelatin was poured 118.0 grams of synthetic clove oil as prepared in Part C. The speed of the stirrer was then adjusted to mix the colloids and the oil. The oil separated into droplets. Two drops of octyl alcohol were added to prevent foaming. The progress of the coacervation was monitored by means of microscopic examination.

The temperature of the mixture was lowered to room temperature, e.g., 24°C. At the higher temperature of 31°C. colloid deposition was observed on the oil droplets. At 24°C. little colloid could be observed in aqueous portions of the mixture. Deposition has ceased. Stirring was continued for 30 minutes, whereupon the reaction mixture was cooled on an ice bath to 4°C. The reaction mixture was maintained at this temperature for 200 minutes. If hardening were desired, 1.0 milliliters of a 25% glutaraldehyde solution in water per gram of gelatin would be added).

The internal phase of the capsules thus formed was approximately 80—90% of the total weight of the capsules.

The capsules thus produced had diameters in the range of from 50 up to 500 microns. They were coated with one of the binders from A or B onto a tobacco sheet material which was shredded and used as a fill in the manufacture of a smoking article.

#### Part E

#### Variation of the Encapsulation of Synthetic Clove Oil

A variation of the encapsulation procedure set forth immediately supra is shown below:

The solution of gum arabic was warmed to 38°C., placed in a Waring blender and stirred. The clove oil prepared in Part C was added gradually while the speed of the blender was being increased until the size of the clove oil droplets was approximately 50—500 microns. The mixture thus formed was poured into a 1000 milliliter beaker containing gelatin, also at 38°C., and was stirred thoroughly. The temperature was then allowed to drop to room temperature and then further decreased to a temperature of 4°C. to 10°C. by means of an ice bath.

It is evident that the tobacco film or filaments can be made from various types and combinations of tobacco. For instance, the tobacco sheet material can be made from relatively expensive tobacco such as Latakia in which it is highly desirable to use all waste because of the high price thereof. So, also, it may be formed of Burley or one or

more scrap or waste cigarette type tobaccos and incorporated in accordance with a particular cigarette manufacturer's formula as if it were natural cigarette tobacco leaves. Any desired formula can thus be maintained in accordance with the demands of a manufacturer's particular brand using one or more types of natural shredded tobacco leaves and admixed desired quantities of shredded capsule containing tobacco film material or filaments, either as a blending or flavoring medium or both or for purposes of bulking.

In the case of the manufacture of cigarettes, according to the present invention, tobacco films are shredded into strands or the film is formed directly into filaments substantially the width of the strands of natural shredded tobacco and of any desired length. In the case of cigars, the capsule containing films are used in large pieces much as long filler tobacco in forming long filler cigars or in smaller pieces for use in the formation of short filler cigars. In all cases the shredded films or filaments or film used in cigarettes and cigars can be handled either manually or by machine in the same manner as natural shredded tobacco leaves or whole leaves or portions thereof. The amount of shredded capsule-containing reconstituted tobacco or pieces of this material employed in a particular blend in cigarettes or cigars, respectively, will vary according to types of tobacco used in the sheet material and the requirements of a particular manufacture.

#### EXAMPLE II

The following is a description of another preferred embodiment of the invention as carried out using a process wherein an aqueous slurry of minute capsules having diameters in the range of 50—500 microns were admixed with shredded natural leaf tobacco.

The capsules and binder materials, when placed among the tobacco shreds, wet them and entangle with them thus securing the capsules against disengaging from the tobacco shreds and separating from a smoking article produced therefrom. Such cigarettes are formed using a wrapper, containing a fill of tobacco extending from one end of the wrapper to the other, and intimately admixed with the tobacco, a plurality of microcapsules each comprising an aromatic volatile synthetic clove oil flavorant. The capsules are homogeneously spaced in contiguous relationship with the tobacco such that as the burning front of the tobacco advances the length of the tobacco article, a concomitant elevation of temperature initiates consecutive rupture of the capsules (1) releasing the volatile synthetic clove oil-containing material which emanates with smoke from the smoking article into the smoker's mouth and (2) yielding a crackling sound audible to the human ear.

## Part A

## Encapsulation of Synthetic Clove Oil

3750 cubic centimeters of synthetic clove oil produced by the procedure of Part C of Example I and 315 grams of gelatin in 6950 cubic centimeters of water was stirred at 200 r.p.m. (six blade axial flow agitator with four evenly spaced baffles) until the particle size was in the range of 50 microns to 500 microns (45 minute stirring). A solution of 315 grams of gum arabic in 6950 cubic centimeters of deionized water was then added. The pH of the resulting mixture was adjusted to 4.5 by the addition of a 10% solution of sodium hydroxide in water. The mixture was slowly cooled on a water bath to a temperature of 38°C. 151.5 cubic centimeters of a 25% solution of glutaraldehyde in water was added. 7.7 grams of sodium benzoate was added. The resulting capsule slurry was stirred for 30 minutes and then filtered using a 20 mesh sieve.

The capsules were then suspended in a mixture of 3 parts gum tragacanth and 17 parts water and the resulting suspension was sprayed onto shredded natural leaf tobacco (using a two fluid pneumatic atomizing nozzle; 1/4 J.A.U.S.S. manufactured by the Spraying Systems, Inc.). The weight ratio of dry capsule to dry shredded tobacco was 0.04:1. The product was used as a fill for cigarettes which, when smoked, released a volatile clove oil flavor and aroma, and yielded a crackling sound audible to the human ear.

## Part B

## Alternate Procedure for Encapsulation of Clove Oil Slurry

10.0 parts by weight of spray dried gum arabic was dissolved in 220.0 parts by weight of deionized water in a beaker equipped with a stirrer. In a separate 250 milliliter Erlenmeyer flask, 10.0 parts by weight of gelatin was dissolved in a 220.0 parts by weight of deionized water.

35.4 parts by weight of  $\beta,\gamma$  - dimethyl -  $\gamma$  - butyrolactone produced by the process of Example III of Canadian Patent 805,934 and 82.6 parts by weight of synthetic clove oil prepared by the process of Part C of Example I were added to the gum arabic solution. The temperature of the mixture was adjusted to 33°C. Thereafter, the solution of gelatin was added with rapid stirring. The reaction mixture was cooled to a temperature of 10°C. on an ice bath while continuously stirred. The mixture was stirred for one hour

at 10°C. Thereafter, 5.0 parts by weight of a 25% solution of glutaraldehyde in water was added to the reaction mass.

The reactant mass was stirred at 10°C. for 8 hours. The resulting capsules had a spherical diameter in the range of from 50 up to 500 microns. The capsules thus formed were not dried but were immediately sprayed onto shredded tobacco. The resulting product was rolled into a cigarette. The cigarette when smoked released a volatile clove oil flavor and aroma and also yielded a crackling sound audible to the human ear.

We are aware of the Customs and Excise Act, 1952, and we make no claim to the use of the invention in contravention of the law.

## WHAT WE CLAIM IS:—

1. An article for smoking comprising a wrapper, tobacco disposed in said wrapper, a plurality of minute rigid-walled, heat-rupturable capsules each comprising an inner phase of volatile flavoring material, said capsules being in intimate contact with said tobacco such that as a burning front of ignited tobacco advances, the capsules are ruptured consecutively by a concomitant elevation in temperature, (1) releasing the volatile flavoring material which is entrained in smoke emanating from the smoking article into a smoker's mouth; and (2) yielding a crackling sound, audible to the human ear.

2. An article as defined in claim 1 wherein said capsules comprise a plurality of micro-capsules each containing an aromatic volatile synthetic clove oil.

3. The article of claim 1 or 2 wherein the micro-capsules possess spherical diameters of from 50 up to 500 microns.

4. The article of claim 3 wherein the heat-rupturable capsules comprise an inner phase of volatile synthetic clove oil and, intimately admixed therewith, an additional tobacco flavor enhancer.

5. The article of claim 4 wherein the weight ratio of dry tobacco to dry capsules is from 1:0.02 up to 1:0.04.

6. An article for smoking substantially as hereinbefore described with reference to Example 1 and Example 2.

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